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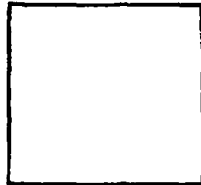
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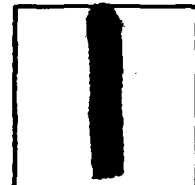
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AN ANALYSIS OF PARAMETERS OF A GAS-DYNAMIC LASER UTILIZING ACETYLENE COMBUSTION

by

M. Syczewski, J. Rutkowski



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AN ANALYSIS OF PARAMETERS OF A GAS-DYNAMIC LASER
UTILIZING ACETYLENE COMBUSTION

Part II. Influence of the composition of gas
mixture on laser parameters

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The effect of various amounts of CO_2 on the operation of a gas-dynamic laser utilizing the heat and products of acetylene combustion was investigated. The optimal amount of CO_2 in the products of combustion was established. It was found that at the optimal amount of CO_2 the radiation energy of laser decreases with increasing amounts of C_2H_2 in substrates, i. e., as the amount of H_2O in products becomes greater. The character of changes of the course of the radiation pulse of laser with time was examined at various values of the pulse energy. The results obtained were discussed.

In reference [1] and in Part I of this investigation [2] the results were given of studies of the effect of gas-dynamic changes on laser parameters with periodic action based on acetylene combustion. In Part I only one composition of gases was employed; the molar ratios were $\text{C}_2\text{H}_2:\text{O}_2:\text{N}_2:\text{CO}_2=1:3.25:14:2.9$. The literature still lacks the results of systematic studies of the effect of gas composition on the parameters. There are some theoretical papers which make use of equations of relaxation kinetics and equations of gas flow dynamics, and analyze the effect of gas composition on the inversion setting of the vibration levels for CO_2 molecules [3], [4]. However, the results of those calculations are more suitable for gas-dynamic lasers of continuous action, where the flow conditions are fixed.

The experimental studies mostly involved the effect of water on laser parameters [5], [6]. These studies do not consider the simultaneous changes of the contents of other components, changes of temperature, pressure and gas-dynamic conditions. The aim of the present work is to extend farther these investigations by including a larger number of the gas mixture parameters.

Experimental conditions

Experiments were conducted on a stand described in [1] with modifications described in Part I of this work [2]. It is a laser of periodic (quasicontinuous) action, in which the gas-dynamic conditions of flow undergo constant changes in the laser pulse. In this work, efforts were made to preserve the constancy of the conditions of runs: the same shape of the nozzle, the same conditions of the supply of gases, and the same (as far as repeatability of adjusting is concerned) optical resonator. These conditions mentioned were not optimized.

In the first series of experiments, the proportion of CO_2 was changed by adding it to the substrate in various amounts instead of N_2 . The initial composition was $\text{C}_2\text{H}_2:3.25\text{O}_2:17\text{N}_2$. CO_2 was added to this composition and N_2 was reduced by the same number of moles. In this way, the same concentration of active components of reaction (acetylene, oxygen) was preserved in the "neutral" gas (nitrogen, carbon dioxide) and care was taken that changes in temperature of the products of combustion would be only slight.

The second series of experiments involved changes in the amounts of acetylene in substrates, and by the same token changes of the amount of water in the products of combustion--with only slight changes of the CO_2 concentration in the products. This was ensured by the addition of varying amounts of CO_2 and C_2H_2 to substrates so as to preserve a constant amount of carbon in the mixture. The composition with the optimal amount of CO_2 from the first series of experiments was used for the initial composition for the second

series. Differences between CO_2 in products between individual runs arose only from different degrees of dissociation of CO_2 . The main measured parameter of laser was the energy of the whole radiation pulse of laser determined with calorimetric recorder which collected radiation from the whole aperture [2].

Simultaneous measurements of the course of intensity of laser radiation and of the pressure in the reaction chamber were also made for characteristic points of the first and the second series. Similarly, as in Part I [2], radiation measurements were done by means of a semiconductor detector (cadmium-mercury telluride) cooled with liquid nitrogen, whose active area was about 1 mm^2 (at the surface of the beam cross-section $\sim 70 \text{ mm}^2$). Since the intensity of radiation at the surface of the cross-section of the laser beam is not uniform, one cannot read the absolute intensity of radiation from the recorded oscillogram, but only its course in time. In addition to these basic series of experiments, a few additional measurements were made, mostly for the purpose of confirming the interpretation of the results. As in [1] and [2], the time variations of the laser pulse radiation and the pressure were recorded on a loop oscillograph as a function of time.

Results and discussion

Tables 1 and 2 give composition of substrates in both series of experiments. The consecutive numbers of compositions correspond to the numbers of points on the graphs discussed below.

Figure 1 presents changes of the radiation energy of laser as a function of the CO_2 concentration in the products of combustion. At the same time the graph shows results of calculation of the temperature of combustion.

Figure 1 shows that the optimal amount of CO_2 in the products of combustion is contained in a rather broad range (17 to 25%). The temperature of combustion products changes, because of different

heat capacities of CO_2 and N_2 . However, this change follows a different course than the change of energy and is nearly linear with the increase of the amount of CO_2 . Different courses of the functions $E = f(\text{CO}_2)$ and $T = f(\text{CO}_2)$ indicate that the lowering of the laser energy is not caused by changes in the gas temperature. This conclusion is confirmed by results of studies by other authors [8] who made investigations at temperatures of 1000-1200 °K. They obtained the curve $E = f(\text{CO}_2)$, similar to ours. Thus, the character of this function is universal for a broad range of temperatures and gas-dynamic conditions.

The obtained results suggest the possibility of raising the chemical efficiency (the degree of utilization of the heat of combustion of acetylene) of this type of lasers by adding a suitable amount of CO_2 above the amount which formed as a result of the combustion reaction.

At an excessive amount of CO_2 we get the so-called concentration quenching of the excited level of antisymmetric vibrations without radiation. Through the collisions of CO_2 molecules between themselves, a quantum of asymmetric vibrations becomes converted into three deformation quanta or one deformation and one symmetric quantum. This is followed by a further radiation-less relaxation from these lower levels, according to the V-T mechanism. Figure 2 indicates that the reduction of the radiation energy of a laser at for high amount of CO_2 occurs as a result of the generalization failure at the beginning of the pulse. In addition to a larger probability of collisions of the excited molecules with nonexcited CO_2 molecules, the reason for interruption of generation at the beginning of the pulse, in the case where there is a large amount of CO_2 in the mixture (oscillogram x), could be a slower combustion reaction rate (a milder increase of pressure). There could also be a low efficiency of the process: $\text{N}_2^* + \text{CO}_2 \rightarrow \text{CO}_2^* + \text{N}_2$, because of the lower N_2 concentration in the gas. The relaxation processes are, however, very complex, because of the fact that they have no linear dependence on temperature (Figure 5).

The graph (Figure 1) shows that at the optimal amount of CO_2 the gas temperature is not yet excessively high, i. e., for this amount of CO_2 one could use a smaller amount of acetylene. This problem was examined more thoroughly in the second series of runs.

Figure 3 shows the course of changes of energy of the radiation pulses of a laser as a function of the amount of acetylene in substrates. The same figure shows changes of the calculated temperature of combustion and of the H_2O and CO_2 contents in the products. The characteristic feature of this series of experiments is a rather sharp change of the temperature of the products when the amount of acetylene changes. One should consider which factor affects the change of laser energy in a decisive manner: the temperature, change in the concentration of CO_2 (concentration changes because of a different degree of dissociation at various temperatures), or change in the concentration of H_2O . To confirm that the temperature is not the decisive factor, we investigated a mixture of ethylene with a similar excess of oxygen as in the case of acetylene. The heat of combustion of ethylene (318 kcal/mole) is similar to the heat of the combustion of acetylene (302 kcal/mole). Hence, the temperature of reaction products in this case will be similar to that of the products of combustion of acetylene--but the amount of water will be twice as great. A mixture containing 4% ethylene--corresponding to the composition a--was burned. If the temperature had the decisive effect upon the radiation energy of the laser we should obtain an amount of energy similar to the value corresponding to the point a (Figure 3). In reality, we obtain about 4 J, which suggests the decisive effect of the water content on the products of combustion.

The effect of varying the amount of CO_2 on the laser energy should also be excluded, since the change in its concentration is small and is contained in a range for which, according to Figure 1, the laser energy is nearly constant and equal to its maximum value.

Figure 4 presents oscillograms which show changes of pressure with time and values of maximum pressure (and thus the gas dynamic conditions of flow) for changes of the amount of acetylene is substrates.

The break in generation, seen on oscillograms for samples d and i at the beginning of the pulse, indicates that the reduction in laser energy occurring with increase of the amount of acetylene is, indeed, caused by the loss of generation at the beginning of the process--and not by lowering of the mean power. The figure shows a considerable change of the maximum pressure on transition of the composition a to compositions d and i. One should consider here whether such an increase of maximum pressure is not the reason for interruption of generation at the beginning of pulse and, by the same token, of the lowering of the laser energy on transition to mixtures rich in acetylene. To consider this problem one should utilize the data from Part I of this work [2].

Taking into consideration that the maximum pressure in sample a is equal about 10 atm, and in samples d and i $p_{\max} = 20$, we note that the ratio p/p_{\max} will change [2] from 0.002 to 0.001. In this range of decrease of the ratio of pressures the value of the radiation energy of the laser should increase. In practice, we observe the contrary effect. It should be acknowledged then that an increase of maximum pressure on transition to mixtures rich in acetylene should not be the reason for the drop in laser energy, and it could rather even mitigate this drop.

The effect of water on laser efficiency is not sufficiently described in the literature and, as indicated by studies of some authors [5], it depends on the gas-dynamic conditions of the laser. For our gas-dynamic conditions, for an increase of the water content from 4.3% to 6.3% the radiation pulse energy of the laser decreased from about 12 to 2 J. The question then arises: how could we change the gas-dynamic conditions so that this drop of energy would be smaller? As we mentioned, relaxation processes

are very complex because of the fact, among others, that the rate of collision deactivation of excited vibrational states by various molecules, particularly of water, changes in a nonlinear manner with a change of temperature.

Figure 5 shows relation between the temperature and the probability of exchange of the vibrational quantum of the CO_2^* molecule into translational energy, as a result of collisions between the molecules CO_2 and H_2O [9]. In the case of collisions $\text{CO}_2^* + \text{CO}_2$, the probability of radiation-less relaxation increases with the temperature rather rapidly. When the temperature changes from normal to 1000° the probability increases by about one order of magnitude. Such a dependence can be justified theoretically and it is characteristic for the majority of gas systems. But the case is different for CO_2 and H_2O molecules. We observe the maximum probability of radiation-less transition of vibrational quanta into translational energy of molecules at about 370°K . When moving either to higher or to lower temperatures, this probability drops rather rapidly.

We should remember that at the time of generation the temperature in resonator cavity (at the outlet of nozzle) changes, hence the probability of radiation-less transitions from vibrational levels of the CO_2 molecule to the ground level changes in an irregular way. In the case of mixtures rich in acetylene, the conditions of the most efficient deactivation process of excited CO_2 molecules by H_2O molecules could exist at the beginning of the laser pulse. From the ratio $p/p_{s\text{max}}$ for particular combustions, one can find [2] the ratio T/T_s . Using the value of this ratio, and from the calculated value of temperature in the combustion chamber T_s , we can calculate the temperature in the resonance cavity T . The obtained temperatures for the oscillograms in Figure 4 are correspondingly: $T_a = 516^\circ\text{K}$, $T_d = 383^\circ\text{K}$, $T_i = 363^\circ\text{K}$. The actual, absolute values of these temperatures can differ from the given ones, but differences between them indicate that the probability of radiation-less relaxation of the excited vibrational states of CO_2 molecules may

differ several fold for compositions (a) and (i). This could be one of the reasons for interruption of generation at the beginning of the pulse.

Changes of the temperature of gases in a resonator cavity depend on the gas flow dynamics. One can change (regulate) the generation by using a different degree of the gas decompression [2]. This explains the fact that it is possible to change the negative effect of water vapor on the laser energy by changing the parameters of the nozzle [5].

Conclusions

On the basis of results obtained in both parts of this work one can state that there is an optimal laser design from a gas dynamic point of view, for each composition and temperature of the combustion products. It is known on the basis of studies and theoretical analyses [4] that the best nozzle profile (of the expanding part) is the so-called logarithmic profile. It can be described by the equation:

$$y = q \ln(ax + c)$$

where:

$$a = c \frac{\operatorname{tg}(\theta_0)}{q}, \quad c = e^{x_0/q}$$

x --distance from critical cross-section;

y --half-height of nozzle at the point x ;

θ_0 --limiting maximum opening angle of the nozzle;

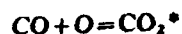
q --a parameter characterizing the degree of nozzle opening.

The higher is value q , the larger is the ratio A/A_{cr} (for a given value of x).

If we take into consideration that the optimal value of the angle θ_0 is about 40° , as found in practice, then the gas-dynamic characteristics of the nozzle will be defined by the parameter q and the width of critical cross-section r_0 .

The results obtained in this work (Part I and Part II) are for the particular values $q = 4$ and $r_0 = 0.35$ mm. The analysis and critical evaluation of these results enable one to make rough estimates of the nozzle design (values of q and r_0) for other gas systems. However, accurate determination of the nozzle dimensions require experimental tests for a particular gas system, because of the necessity of taking into account a number of interdependent parameters. At the existing nozzle profile, the usually modification consists of a change of the size of the critical cross-section (r_0).

It is necessary to state that our way of carrying experiments, which involves the opening of the gas flow channel before the completion of combustion process. It differs from the experiments performed by the cited authors. It cannot be excluded in our case that the first period of the laser radiation pulse in the area where the pressure still increases (where the reaction still develops) arises as a result of chemical pumping [10]. Each oscillogram shows a short pulse at the beginning whose intensity decreases or even vanishes near the pressure maximum. Then, the intensity of laser radiation begins to increase again, and this second, main part of pulse remains more stable up to the end. It is quite possible, therefore, that in the first period the laser operates as a chemical laser, according to the reaction



When combustion in the chamber nears completion, the actual period gas-dynamic laser operating period begins.

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TABLE 1

Composition of gases (substrates) in the first series of experiments

(1) Oznaczenie zestawu	(2) Skład mieszaniny gazowej [mm Hg, %]				(3) średnia energia lasera [J]
	C ₂ H ₂	O ₂	CO ₂	N ₂	
1	64 4,68	208 15,20	— —	1096 80,12	5,8
2	—	—	11 0,80	1085 79,31	6,5
3	—	—	22 1,61	1074 78,51	7,3
4	—	—	33 2,41	1063 77,71	7,9
5	—	—	34 3,22	1052 76,90	8,6
6	—	—	55 4,02	1041 76,10	8,9
7	—	—	110 8,04	986 72,08	12,0
8	—	—	165 12,06	931 68,06	11,74
9	—	—	220 16,08	876 64,04	11,5
10	—	—	275 20,10	821 60,02	10,3
11	—	—	330 24,12	766 56,00	6,2
12	—	—	385 28,14	711 51,98	5,2
13	—	—	440 32,16	656 47,96	3,6
14	—	—	495 36,18	601 43,94	2,8

Key: 1 - Designation of set; 2 - Composition of gas mixture (mm Hg), (%); 3 - Mean energy of laser (J)

TABLE 2
Composition of gases (substrates) in the second
series of experiments

(1) Oznaczenie zestawu	(2) Skład mieszaniny gazowej [mm Hg]. [%]				(3) Średnia energia lasera [J]
	C ₂ H ₂	O ₂	CO ₂	(Hg)	
a	55 4,08	179 13,28	183 13,58	935 69,06	10,8
b	60 4,42	195 14,35	173 12,73	931 68,50	10,94
g	64 4,68	208 15,20	165 12,06	931 68,50	9,22
c	70 5,06	228 16,50	153 11,07	931 67,37	8,4
d	75 5,38	244 17,52	143 10,26	931 66,83	5,3
e	80 5,70	260 18,52	133 9,47	931 66,31	3,6
f	85 6,01	276 19,50	123 8,69	931 65,80	3,8
g	90 6,31	292 20,48	113 7,92	931 65,29	3,50
h	95 6,61	309 21,49	103 7,16	931 64,74	3,55
i	100 6,90	325 22,43	93 6,42	931 64,25	1,9

Key: 1 - Designation of set; 2 - Composition of gas mixture (mm Hg), (%); 3 - Mean energy of laser (J).

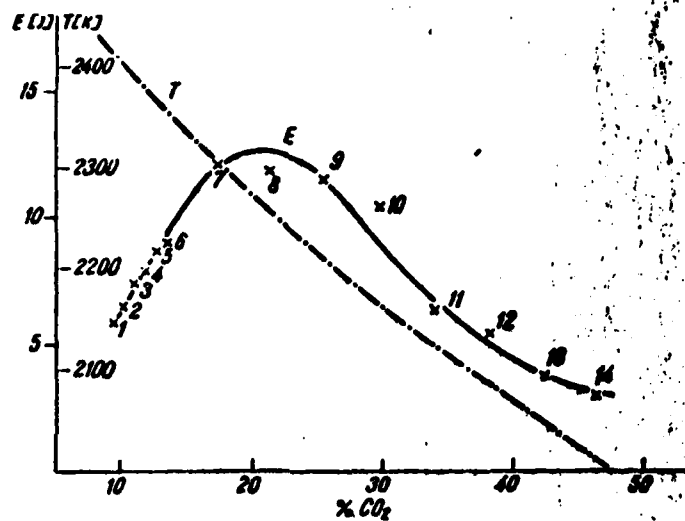
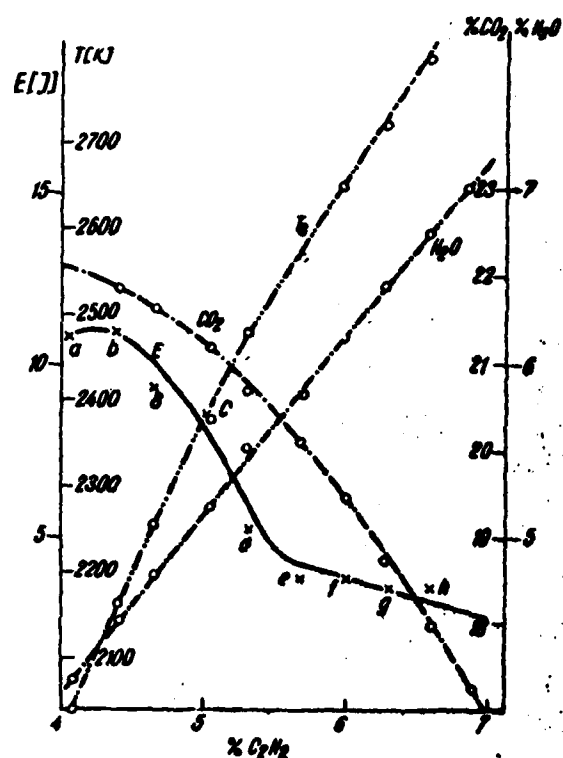


Figure 1. Dependence of the radiation energy E of a laser (in the whole pulse) and of the temperature of combustion T on CO_2 concentration in combustion products; each point represents value of at least three measurements.



Figure 2. Oscillograms of the intensity of laser radiation and of pressure for three different compositions of gases (according to Table 1); division of time scale - 0.05 sec.
 a - composition No. 1; b - composition No. 2; c - Composition No. 3

Figure 3. Changes of the radiation energy of laser impulse E , temperature of combustion T , contents of CO_2 and of H_2O in the products--as a function of the contents of acetylene in substrates (sets denoted according to Table 2).



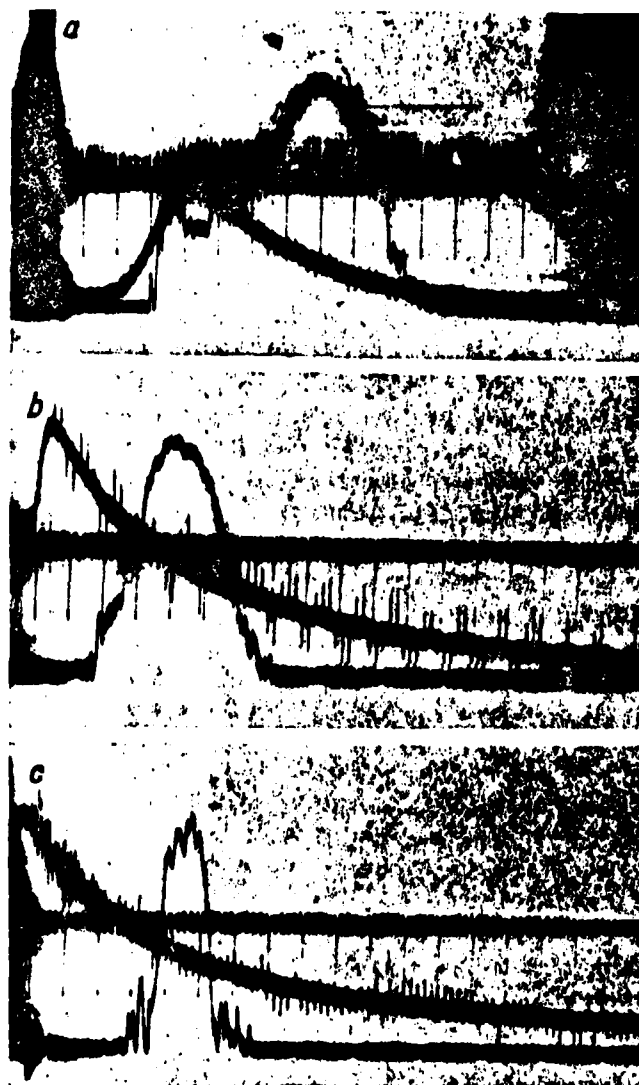


Figure 4. Oscillograms of the intensity of laser radiation and of pressure for three mixtures with different contents of acetylene (according to Table 2); division of time scale - 0.05 sec

a - composition a; b - composition d; c - composition i

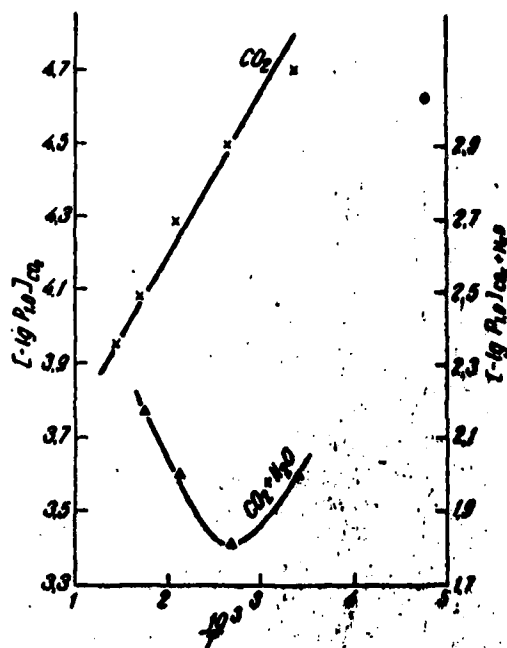


Figure 5. Dependence of the probability P for radiation-less relaxation of excited vibrational states of the molecule CO_2^* , as a result of collisions with CO_2 and H_2O on temperature-

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